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13. ABSTRACT (Maximum 200 words) Instrumented high velocity impact recovery experiments were performed to study solid state reactions in mixtures of nickel and aluminum powders during the passage of shock waves. "Excess pressure" due to the release of energy from the exothermic reaction was measured for a range of impact velocities. An impact velocity threshold for the initiation of reaction at the reflected shock front was established and this threshold value was found to be dependent on the particle size of the powder. Microscopic examination of the recovered specimens show features which corroborate hypothesized extent of reaction based on pressure measurements.					
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SHOCK WAVE INTERACTIONS WITH EXOTHERMIC MIXTURES

INTRODUCTION

Until as recently as 1987, the possibility of chemical reaction at the shock front was speculative. Thadhani [1] has written an exhaustive review of the subject of shock-induced chemical reactions which also covers the subject of chemical reactions at the shock front. This investigation is based on the premise that the examination of the post-shock recovery specimen alone will not establish when the reaction took place. It is clear that understanding the kinetics of such reactions, which are obviously beyond the realm of diffusion mechanisms, will require measurements of changes at the shock front. The energy released by exothermic reactions is expected to manifest itself as pressure and temperature. We chose to attempt to make time-resolved pressure measurements when a green compact of a suitable mixture of powders of nickel and aluminum is subjected to planar high velocity impact.

EXPERIMENTS

The details of the recovery fixture, powder gun assembly, velocity and pressure measurement techniques, and the preparation of specimens have been described in previous publications [2-4]. Gauges located in front of the specimen to measure the incident wave on the specimen did not survive to produce

reliable measurements. Manganin gauge epoxied to the back of the rear cover disc survived for about a microsecond and produced reproducible records during the half a microsecond between the reflection of the compression wave from the rear cover disc and the arrival of release wave. The recording digitizing oscilloscope gave a data point every 50 nanoseconds.

Green compacts of 55% of the theoretical density of solid mixture of Ni and Al powders were impacted by a 304 stainless steel flyer plate at the front surface of a lexan projectile driven to a series of velocities in a 19mm powder gun. The size of the Al powder was 20 μm in all the experiments; two series of experiments, one with Ni powder of 3-7 μm and another with <3 μm size were performed. Experiments with pure Al powder compact and Ni powder compacts were also performed to compare the calculated peak reflected pressure values with the measured values and validate the reliability of the measurement technique. A 2.6:1 Ni to Al atomic weight percent ratio was maintained to keep the impedance of the fully densified mixture lower than that of the 304 SS in order for the wave reflected from the rear cover disc to be compressive. After the experiment, portions of the specimen, strongly

adhered to the rear cover disc, were recovered and subjected to metallographic examination, usually in the polished and unetched condition. Nondispersive Energy X-ray spectroscopy in the Scanning Electron Microscope helped identify the reaction products qualitatively.

RESULTS

Figure 1 shows a typical pressure-time record. The impact velocity was 1075 m/s. The predicted value is the reflected peak pressure at the interface of a fully dense inert mixture of Ni and Al in the ratio of 2.6:1 by atomic weight

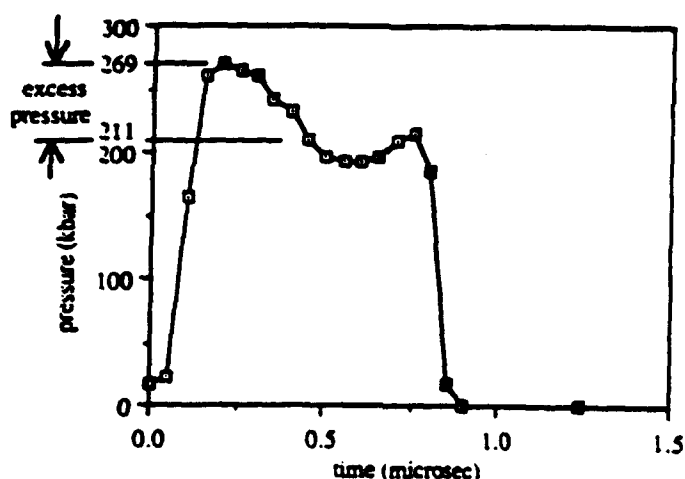


Figure 1. Typical Pressure-Time data for an impact experiment. (1075 m/s. 3-7 μ m Ni powder)

and 304 SS rear cover plate. The shock properties were calculated from published data for Ni and Al and on the bases of simple mixture theory and the assumption of complete densification by the passage of the incident shock wave

through the specimen. Temperature effects due to the heating of the specimen were not considered. In a series of experiments with increasing impact velocity, a threshold velocity, below which the measured peak pressure was slightly lower than the predicted peak pressure and above which the measured value was much higher than the predicted value, was observed. Above the

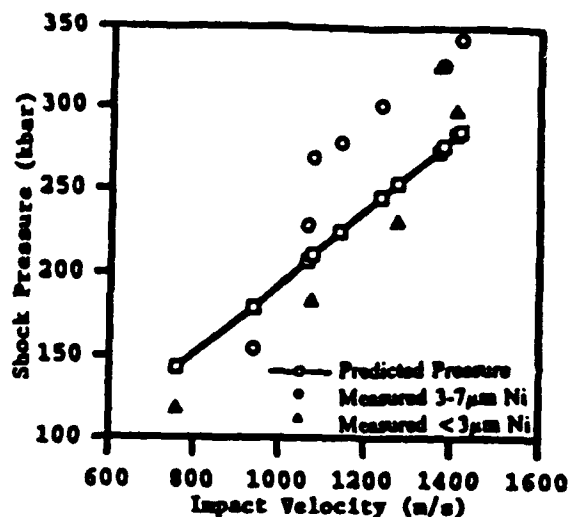


Figure 2. Measured reflected shock pressure vs impact velocity for two series of experiments.

threshold the "excess pressure" was about the same irrespective of the impact velocity. In another series of similar experiments with a smaller size Ni powder in the mixture, the threshold at which the excess pressure was observed was shifted to a higher impact velocity. Results of both series of experiments are summarized in Figure 2.

At low velocities of impact, the recovered specimens did not show any sign of compound formation. At high enough but lower than the threshold impact velocity, specimens revealed

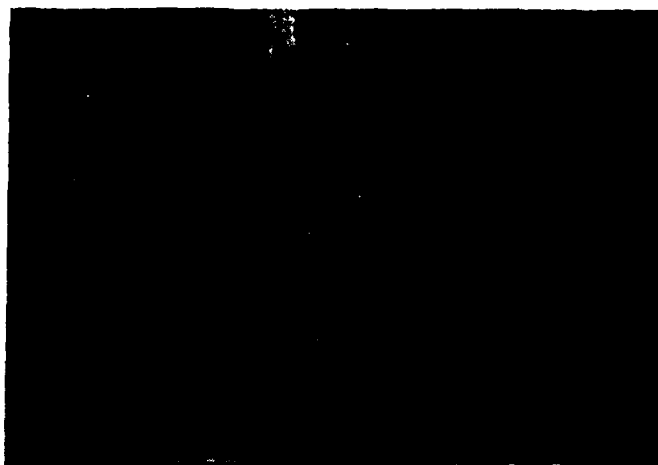


Figure 3. Photomicrograph of a sub-threshold recovery specimen. No "excess pressure" was evident in this experiment. (1075 m/s. $< \mu\text{m Ni}$) 300X

regions of reaction products. Figure 3 is a photo micrograph of a recovered specimen with regions of unreacted Al surrounded by rings of light and gray regions of Al + Ni compound and unreacted Ni. Above the threshold but at the lower velocity region all the aluminum had combined with nickel but Ni-rich and Al-free regions are still



Figure 4. Scanning electron micrograph of a post-threshold specimen. Melting of the product is indicated. (1380 m/s. $3/7 \mu\text{m Ni}$) 750X

present. At higher velocities reaction products were found all over with a range of Al-Ni ratios. Figure 4 shows regions of compound melting.

DISCUSSION

At impact velocities near but lower than threshold velocity, consistent with our previously reported results [2] on the Sn-S system, reaction products were found but the measured peak pressure was lower than the predicted value. Heating by the shock is sufficient to melt the aluminum in the mixture thus making any reaction diffusion-controlled and the post-shock reaction proceeds but is arrested by the massive quenching by the fixture. Above the threshold, the "excess pressure" recorded is reproducible and much larger than the possible experimental errors in measurement (3%). Exothermic reaction at the reflected shock front is a definite possibility. Obviously, the extent of reaction in the recovered specimen is more than what could have occurred during the passage of the reflected shock wave because the reaction could continue by diffusion in the post-shock specimen until, as explained before, the fixture would arrest the reaction. It is reasonable to expect the same amount of reaction in the reflected shock front irrespective of the impact velocity especially if one considers the small range of impact velocities in this investigation. But at higher impact velocities, the specimen is heated to a greater extent and the diffusion controlled reaction proceeds further. Small regions of what is considered to be molten and solidified reaction products

could be found only in specimens impacted at the high velocities. The pressure-time traces at these velocities also showed greater drops after the peak value of pressure.

In an experiment in which the back cover plate of 304 stainless steel was replaced by lexan whose impedance is less than that of the dense mixture of Ni + Al. The recovered specimen did not show any sign of reaction even though the impact velocity was about 1300 m/s. Complete pore collapse and a certain similarity with recovered specimens impacted at velocities lower than the threshold value were observed; this would lead one to believe that heating by pore collapse alone may not be sufficient to trigger and sustain the reaction by shock wave. Unfortunately, a pressure-time trace was not obtained in this experiment. Nevertheless, the possibility of the incident shock densifying and conditioning the mixture such that the reaction took place only in the reflected shock is indicated. It would have been preferable to conduct an experiment at high enough impact velocity which corresponded to the energy levels of the reflected shock in the specimen. Currently we do not have facilities in our laboratory for that.

The effect of the smaller Ni particle size in the second series of experiments was to shift the threshold velocity to a higher value. The impact velocity for the ultrafast reaction thresholds of the two Ni-Al mixtures appears to be inversely related to the particle size. This may be a clue to a physical description of the ultrafast

reaction mechanisms. The mixture constituents have different impedances and it is not realistic to assume that the particle velocities are immediately in equilibrium during and just behind the shock front. So a relative particle velocity may exist between the constituents [5]. In fact this relative particle velocity may be greatly increased during the reflected shock front where it is possible for the constituents to be moving in opposite directions for a short period [6]. This relative particle velocity will be equilibrated by inter-particle shear which is also inversely related to the particle size. A recent Eulerian model of this process shows that this equilibration takes place in about 100 nanoseconds [6], i.e., in and just behind the reflected shock front. This inter-particle shear will rapidly expose fresh reaction surfaces and cause extreme mass mixing. Dremin and Breusov [7] have indicated that reaction nucleation and propagation may occur under these conditions within these time constraints. If a critical value in inter-particle shear must be overcome for these ultrafast reactions to occur, they will occur at different impact velocities for different particle size mixtures.

CONCLUSIONS

Time-resolved measurements of the reflected shock pressure profile through exothermic mixtures have yielded measured peak pressures in excess of those predicted for the inert systems. This excess pressure is attributed to the heat released by ultrafast exothermic reactions at the shock front. Ultrafast reaction thresholds have been

observed and the reactions have been observed to occur in the reflected shock front. The reaction takes place in about 100 nanosecond. The occurrence of ultrafast reaction thresholds has been corroborated through material analysis of the recovered specimens. Assuming that the ultrafast reaction product is NiAl_3 , Hugoniot calculations which account for the heat of reaction show that the excess pressure represents a nearly complete reaction. A change in the size of the nickel particles used in the Ni-Al mixture resulted in a substantial shift in the ultrafast reaction threshold. This hints at a physical description of the reaction mechanism. The ultrafast reaction mechanism may be related to interparticle shear which is necessary to equilibrate relative particle velocities within and just behind the shock front and may rapidly expose fresh reaction surfaces for ultrafast reactions.

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